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Tuning the static spin-stripe phase and superconductivity in $La_{2-x}Ba_xCuO_4$ ($x = 1/8$) by hydrostatic pressure

Z Guguchia[1,4], A Maisuradze[1,2], G Ghambashidze[3], R Khasanov[2], A Shengelaya[3] and H Keller[1]

[1] Physik-Institut der Universität Zürich, Winterthurerstrasse 190, CH-8057 Zürich, Switzerland
[2] Laboratory for Muon Spin Spectroscopy, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland
[3] Department of Physics, Tbilisi State University, Chavchavadze 3, GE-0128 Tbilisi, Georgia
E-mail: zurabgug@physik.uzh.ch

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Abstract. Magnetization and muon spin rotation experiments were performed in $La_{2-x}Ba_xCuO_4$ ($x = 1/8$) as a function of hydrostatic pressure up to $p \simeq 2.2$ GPa. It was found that the magnetic volume fraction of the static stripe phase strongly decreases linearly with pressure, while the superconducting volume fraction increases by the same amount. This demonstrates competition between bulk superconductivity and static magnetic order in the stripe phase of $La_{1.875}Ba_{0.125}CuO_4$ and that these phenomena occur in mutually exclusive spatial regions. The present results also reveal that the static spin-stripe phase still exists at pressures, where the long-range low-temperature tetragonal (LTT) structure is completely suppressed. This indicates that the long-range LTT structure is not necessary for stabilizing the static spin order in $La_{1.875}Ba_{0.125}CuO_4$.

[4] Author to whom any correspondence should be addressed.

$La_{2-x}Ba_xCuO_4$ (LBCO) was the first cuprate in which high-T_c superconductivity was discovered [1]. The undoped parent compound is an antiferromagnetic (AFM) insulator. The replacement of La^{3+} by Ba^{2+} ions, through which holes are doped into the CuO_2 planes, causes the destruction of AFM order and superconductivity appears at $x = 0.06$. Subsequent investigations showed that there exists a sharp dip in the T_c–x phase diagram, indicating that bulk superconductivity is greatly suppressed in a narrow range around a particular doping concentration $x = 1/8$ in LBCO [2]. This suppression of T_c has attracted a great deal of attention and is known in the literature as the 1/8 anomaly (see e.g. [3, 4]). Later a similar anomaly was also observed in rare earth doped $La_{2-x}Sr_xCuO_4$. Studies of the crystal structure clarified that the LBCO system undergoes at $x = 1/8$ a first-order structural phase transition from a low-temperature orthorhombic (LTO) to a low-temperature tetragonal (LTT) phase with decreasing temperature [5]. Since the structural transition to the LTT phase appears near the Ba concentration x where the strong decrease of T_c occurs, it has been suggested that there is a correlation between the appearance of the LTT phase and the suppression of superconductivity [5]. Muon spin rotation (μSR) experiments detected the appearance of static magnetic order below ~ 30 K in $La_{1.875}Ba_{0.125}CuO_4$ (LBCO-1/8) [6].

The discovery of elastic superlattice peaks in $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ by neutron diffraction provided evidence of two-dimensional (2D) charge and spin order, which was explained in terms of a stripe model where charge-carrier poor AFM regions are separated by one-dimensional stripes of charge carrier-rich regions [7, 8]. The presence of stripe-like charge and spin density ordering is believed to be responsible for the anomalous suppression of superconductivity around $x = 1/8$ in cuprates [7, 8]. The existence of stripes in $La_{1.85}Sr_{0.15}CuO_4$ and $Bi_2Sr_2CaCu_2O_{8+y}$ has also been demonstrated by extended x-ray absorption fine structure experiments which allow to probe the local structure near a selected atomic site [9, 10].

The fascinating issue of charge and spin stripes in cuprate superconductors has attracted a lot of attention for many years (see e.g. [3, 4]). Experimental results and theoretical considerations show that the modulations of the lattice and of the charge and spin density appear to be both ubiquitous in the cuprates and intimately tied up with the physics of these materials [3, 4]. However, the role of stripes for superconductivity in cuprates is still unclear at present. Therefore, it is important to find an external control parameter which allows to tune structural and electronic properties of the cuprates and study the relation between superconductivity and stripe order. It is known that upon applying hydrostatic pressure both the LTT and LTO structural phase transition in LBCO-1/8 are suppressed completely at the critical pressure $p_c \approx 1.85$ GPa, and superconductivity is enhanced [11–14]. The magnetic order related to stripe formation was previously studied under pressure, but only below p_c [15]. Hence, it is not known how the static spin-stripe order changes across p_c.

Here, we report studies of superconductivity and stripe magnetic order in LBCO-1/8 under hydrostatic pressure up to $p \simeq 2.2$ GPa by magnetization and μSR experiments. It was observed that the transition temperature of the stripe magnetic order and the size of the ordered moment are not significantly changed by pressure. But the volume fraction of the magnetic phase significantly decreases and simultaneously the superconducting (SC) volume fraction increases with increasing pressure. This indicates that magnetic regions in the sample are converted to SC regions with increasing pressure, providing evidence for a competition between superconductivity and static magnetic order in the stripe phase of LBCO-1/8. It was also demonstrated that the spin-stripe order still exists at pressures, where the LTT phase is suppressed.

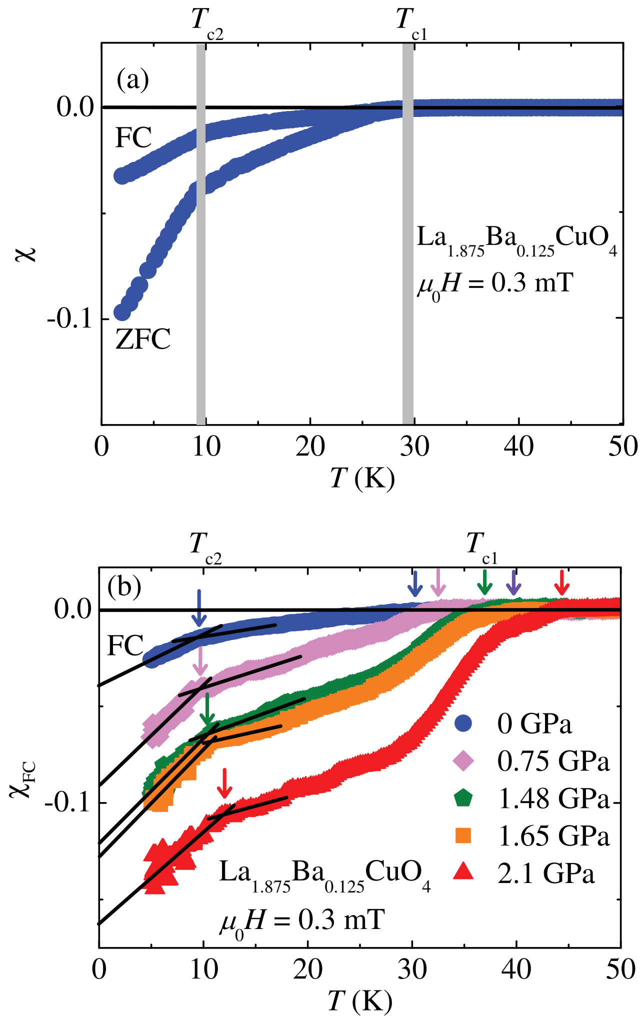

Figure 1. Temperature dependence of the magnetic susceptibility of LBCO-1/8 measured at ambient pressure without pressure cell (a) and at various applied hydrostatic pressures (b) in a magnetic field of $\mu_0 H = 0.3$ mT. The vertical grey lines and the arrows denote the SC transition temperatures T_{c1} and T_{c2} (see text for an explanation).

A polycrystalline sample of $La_{2-x}Ba_xCuO_4$ with $x = 1/8$ was prepared by the conventional solid-state method. The single-phase character of the sample was checked by powder x-ray diffraction.

The magnetic susceptibility was measured under pressures up to 2.1 GPa by a SQUID magnetometer (*Quantum Design* MPMS-XL). Pressures were generated using a diamond anvil cell (DAC) [16] filled with Daphne oil which served as a pressure-transmitting medium. The pressure at low temperatures was determined by the pressure dependence of the SC transition temperature of Pb. The temperature dependence of the zero-field-cooled (ZFC) and field-cooled (FC) magnetic susceptibility, χ_{ZFC} and χ_{FC}, respectively, for LBCO-1/8 in a magnetic field of $\mu_0 H = 0.3$ mT is shown in figure 1(a). The diamagnetic susceptibility exhibits a two-step SC transition. The first transition with an onset at $T_{c1} \approx 30$ K corresponds to only about 4% volume fraction of superconductivity estimated from ZFC magnetization at 10 K.

The second SC transition is observed at $T_{c2} \approx 10\,\mathrm{K}$, with a larger diamagnetic response. However, the volume fraction of the low temperature SC phase is still small at ambient pressure and amounts to about 10% of full shielding at 2 K. A two-step SC transition, starting at around 30 K with a weak diamagnetic response was observed previously in polycrystalline LBCO-1/8 [2, 11]. It was explained as some kind of filamentary superconductivity due to the presence of a very small fraction of the LTO phase. Recent detailed transport and susceptibility measurements in single crystal of LBCO-1/8 provided evidence of the intrinsic nature of the observed two-step SC transition [17]. It was found that a SC transition at higher temperature T_{c1} is present when the magnetic field is applied perpendicular to the CuO_2 planes. The SC transition at low temperature T_{c2} is more pronounced when the magnetic field is parallel to the planes ($H \parallel ab$). The authors interpreted the transition at T_{c1} as due to the development of 2D superconductivity in the CuO_2 planes, while the interlayer Josephson coupling is frustrated by static stripes. A transition to a three-dimensional SC phase takes place at much lower temperature $T_{c2} \ll T_{c1}$, reflected as a strong increase of diamagnetism below T_{c2} for $H \parallel ab$. For polycrystalline samples with random orientation of grains these two temperatures will result in two SC transitions as observed in present experiments (see figure 1(a)).

We studied the SC transition in LBCO-1/8 as a function of hydrostatic pressure. Measurements were performed in the FC mode at 0.3 mT, which was set constant during the measurements at all pressures in order to avoid a variation of the applied field during the measurements with different pressures. Figure 1(b) shows the temperature dependence of χ_{FC} for different pressures after substraction of the background signal from the empty pressure cell. A two-step SC transition is observed at all pressures, while at higher pressure ($p > 1.6\,\mathrm{GPa}$) even a three-step SC transition is visible. The reason for this is not clear at present. Further investigations, in particular on single crystals, are needed to clarify this issue. It was found that T_{c2} increases only slightly with pressure from 10 to about 12 K at the maximal pressure applied in our experiments ($p = 2.1\,\mathrm{GPa}$). On the other hand, T_{c1} shows a significant increase with a rate of $6.2\,\mathrm{K\,GPa^{-1}}$. It is interesting that the volume fraction of the corresponding SC phase is also strongly enhanced with applied pressure (see figure 1(b)). These results are in agreement with previous studies showing that superconductivity in LBCO-1/8 is largely enhanced by applying pressure [11, 13, 18].

It is interesting to explore the pressure effect on spin order in the stripe phase and its relation to superconductivity. It is also of great interest to study the relation between static magnetism and the LTT phase in LBCO-1/8. However, to the best of our knowledge magnetism in LBCO-1/8 was studied only at low pressures [15] where the LTT phase is still present. In order to answer this question we performed zero-field (ZF) μSR experiments in LBCO-1/8 at ambient and various hydrostatic pressures, including pressures where the long-range LTT structure is suppressed. ZF μSR is a powerful tool to investigate microscopic magnetic properties of solids without applying an external magnetic field. It is especially suitable for the study of weak magnetic order, since the positive muon is an extremely sensitive local probe which is able to detect small internal magnetic fields and ordered volume fractions.

The ZF μSR experiments were carried out at the μE1 beam line at the Paul Scherrer Institute, Switzerland. Pressures up to 2.2 GPa were generated in a double wall piston-cylinder type of cell made of MP35N material, especially designed to perform μSR experiments under pressure [19]. As a pressure transmitting medium Daphne oil was used. The pressure was measured by tracking the SC transition of a very small indium plate. The μSR time spectra were analysed using the free software package MUSRFIT [20].

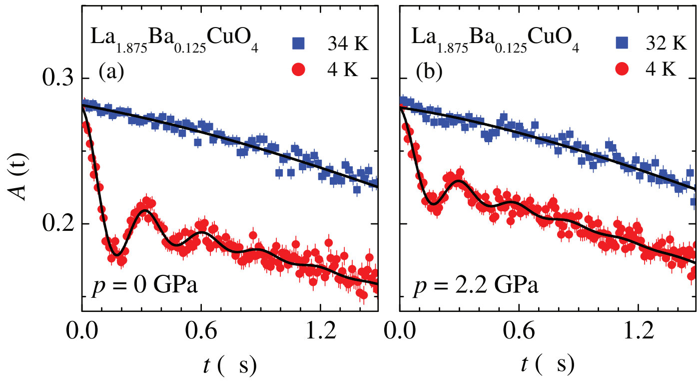

Figure 2. ZF μSR signal $A(t)$ of LBCO-1/8 measured at $p = 0\,\mathrm{GPa}$ (a), and 2.2 GPa (b), recorded for two different temperatures: $T = 4\,\mathrm{K}$ (circles) and $T = 32\,\mathrm{K}$ (squares). The solid lines represent fits to the data by means of equation (1).

Figure 2 shows representative ZF μSR time spectra for a polycrystalline LBCO-1/8 sample at ambient and at maximum applied pressure $p = 2.2\,\mathrm{GPa}$, respectively. Below $T \approx 30\,\mathrm{K}$ damped oscillations due to muon-spin precession in local magnetic fields are observed, indicating static spin-stripe order [6, 21].

A substantial fraction of the μSR asymmetry signal originates from muons stopping in the MP35N pressure cell surrounding the sample. Therefore, the μSR data in the whole temperature range were analysed by decomposing the signal into a contribution of the sample and a contribution of the pressure cell:

$$A(t) = A_{\mathrm{S}}(0)\,P_{\mathrm{S}}(t) + A_{\mathrm{PC}}(0)\,P_{\mathrm{PC}}(t), \tag{1}$$

where $A_{\mathrm{S}}(0)$ and $A_{\mathrm{PC}}(0)$ are the initial asymmetries and $P_{\mathrm{S}}(t)$ and $P_{\mathrm{PC}}(t)$ are the muon-spin polarizations belonging to the sample and the pressure cell, respectively. The pressure cell signal was analysed by a damped Kubo–Toyabe function [19]. The response of the sample consists of a magnetic and a nonmagnetic contribution

$$P_{\mathrm{S}}(t) = V_{\mathrm{m}}\left[\frac{2}{3}\,\mathrm{e}^{-\lambda_{\mathrm{T}}t}\,J_0(\gamma_\mu B_\mu t) + \frac{1}{3}\,\mathrm{e}^{-\lambda_{\mathrm{L}}t}\right] + (1 - V_{\mathrm{m}})\,\mathrm{e}^{-\lambda_{\mathrm{nm}}t}. \tag{2}$$

Here, V_{m} denotes the relative volume of the magnetic fraction, and $\gamma_\mu/(2\pi) \simeq 135.5\,\mathrm{MHz\,T^{-1}}$ is the muon gyromagnetic ratio. B_μ is the average internal magnetic field at the muon site. λ_{T} and λ_{L} are the depolarization rates representing the transversal and the longitudinal relaxing components of the magnetic parts of the sample. J_0 is the zeroth-order Bessel function of the first kind. This is characteristic for an incommensurate spin density wave and has been observed in cuprates with static spin stripe order [21]. λ_{nm} is the relaxation rate of the nonmagnetic part of the sample. The total initial asymmetry $A_{\mathrm{tot}} = A_{\mathrm{S}}(0) + A_{\mathrm{PC}}(0) \simeq 0.285$ is a temperature independent constant. A typical fraction of muons stopped in the sample was $A_{\mathrm{S}}(0)/A_{\mathrm{tot}} \simeq 0.50(5)$ which was assumed to be temperature independent in the analysis.

The temperature dependence of B_μ for different pressures is shown in figure 3(a). The solid curves in figure 3(a) are fits of the data to the power law $B_\mu(T) = B_\mu(0)[1-(T/T_{\mathrm{so}})^\gamma]^\delta$, where $B_\mu(0)$ is the zero-temperature value of B_μ. γ and δ are phenomenological exponents. The values of the spin ordering temperature $T_{\mathrm{so}} \simeq 30\,\mathrm{K}$ and $B_\mu(0) \simeq 25\,\mathrm{mT}$ at ambient pressure are in good agreement with the values of a previous μSR study [15, 21]. As evident from

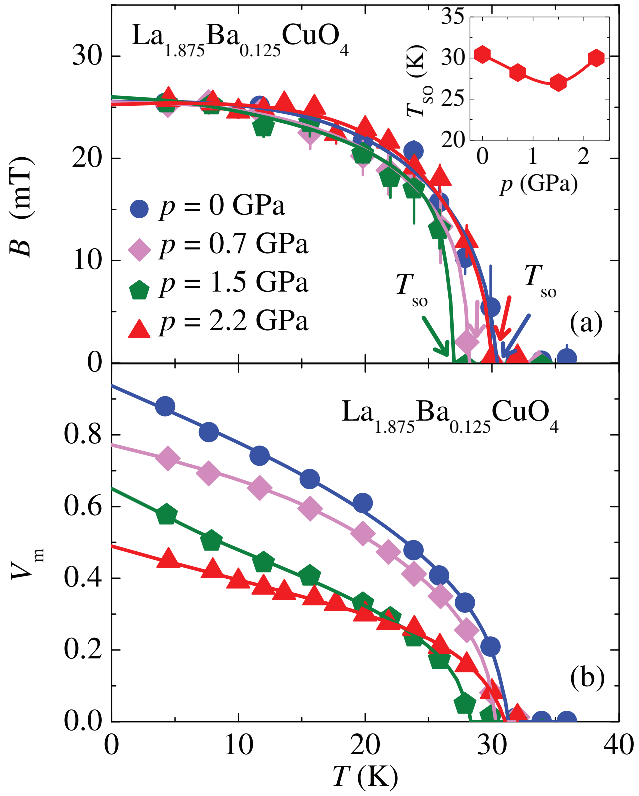

Figure 3. (a) Temperature dependence of the average internal magnetic field B_μ at the muon site of LBCO-1/8 recorded at various applied pressures. The solid lines represent fits of the data to the power law described in the text. The arrows mark the transition temperatures for the static spin-stripe order T_{so}. The inset shows T_{so} as a function of pressure p. (b) The temperature dependence of the magnetic volume fraction V_m in LBCO-1/8 at ambient and various hydrostatic pressures. The solid lines are fits of the data to a similar empirical power law as used for $B_\mu(T)$ in (a).

figure 3(a) the internal magnetic field $B_\mu(0)$ is almost pressure independent. This indicates that the ordered magnetic moment of the static stripe phase does not depend on applied pressure. Also T_{so} changes only slightly with pressure as shown in the inset of figure 3(a). In the pressure range of $p = 0 - 2.2$ GPa, $T_{so}(p)$ varies only between 30 and 27 K with a shallow minimum at $p \simeq 1.5$ GPa.

It is important to note that both the LTT and LTO structural phase transition are suppressed at $p_c = 1.85$ GPa [14]. Therefore, the present results demonstrate that the spin order due to static stripes still exists at $p = 2.2$ GPa, where the LTT phase is already suppressed. Recent high pressure x-ray diffraction experiments showed that also the charge order of the stripe phase survives above p_c in LBCO-1/8 [14]. Combining these results, one may conclude that both charge and spin order, and consequently the static stripe phase itself, still exist at pressures where the LTT phase is suppressed.

Here the question arises: What is the effect of pressure on the stripe order in LBCO-1/8? In agreement with the previous low-pressure μSR results [15], it was found that it is the

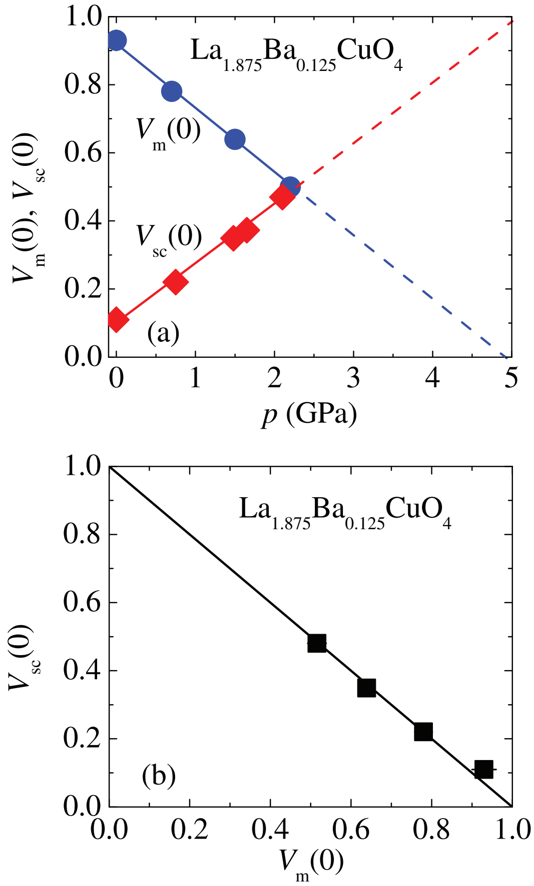

Figure 4. (a) The pressure dependence of the zero-temperature limit of the magnetic and the SC volume fractions, $V_m(0)$ and $V_{sc}(0)$, respectively, of LBCO-1/8. Solid lines are linear fits to the data. (b) $V_{sc}(0)$ versus $V_m(0)$. The solid straight line is drawn between a hypothetical situation of a fully magnetic ($V_m(0)$ = 1) and a fully SC state ($V_{sc}(0)$ = 1).

magnetic volume fraction V_m which is significantly suppressed by pressure. μSR can determine the ordered volume fraction and is thus a particularly powerful tool to study inhomogeneous magnetism in materials. Figure 3(b) shows the temperature dependence of V_m at various pressures. V_m increases progressively below T_{so} with decreasing temperature and acquires nearly 100% at ambient pressure at the base temperature [6]. An important result is that at low temperature V_m significantly decreases with increasing pressure (see figure 3(b)). This means that with increasing pressure an increasingly large part of the sample remains in the nonmagnetic state down to the lowest temperatures.

In order to compare the influence of pressure on the SC and magnetic properties of LBCO-1/8, the pressure dependences of the zero-temperature limit of the magnetic volume fraction $V_m(0)$ and the SC volume fraction $V_{sc}(0) = -\chi_{ZFC}(0)$ are plotted in figure 4(a) [5]. Note that $V_m(0)$

[5] As mentioned in the text we measured only χ_{FC} under high pressure. The difference $\chi_{ZFC}(0) - \chi_{FC}(0)$ was obtained from the measurements at ambient pressure (see figure 1(a)). By assuming that this difference does not change with pressure, the value of χ_{ZFC} at $T = 0$ K was determined at different applied pressures.

linearly decreases with pressure to approximately 50 % at $p = 2.2$ GPa. A linear extrapolation of $V_{\rm m}(0)$ to higher pressures shows that the magnetic volume fraction should be completely suppressed at $p \approx 5$ GPa. It would be interesting to check this prediction at higher pressures by either μSR or neutron-scattering experiments. It is evident from figure 4(a) that the decrease of $V_{\rm m}(0)$ is followed by an increase of the SC volume fraction $V_{\rm sc}(0)$. In figure 4(b) we plot $V_{\rm sc}(0)$ as a function of $V_{\rm m}(0)$. The solid straight line is drawn between a hypothetical situation of a fully magnetic ($V_{\rm m}(0) = 1$) and a fully SC state ($V_{\rm sc}(0) = 1$). Remarkably, the experimental data lie on this solid straight line. Thus, the sum of the SC and magnetic volume fractions is constant and is close to one. This strongly suggests that superconductivity does not exist in those regions where static magnetism is present. Thus, superconductivity most likely develops in those areas of the sample which are nonmagnetic down to the lowest temperatures. The latter implies that in LBCO-1/8 magnetism and superconductivity are competing order parameters. It is interesting to note that a similar scaling was found between the superfluid density and the magnetic volume fraction in the related compound $La_{1.85-y}Eu_ySr_{0.15}CuO_4$ [22]. The tuning of the magnetic and SC properties was realized by rare-earth doping.

To summarize, magnetism and superconductivity were studied in LBCO-1/8 by means of magnetization and μSR experiments as a function of pressure up to $p \simeq 2.2$ GPa. It was demonstrated that the static spin-stripe order still exist at pressures, where the long-range LTT structure is suppressed. This suggests that the long-range LTT phase is not essential for the existence of stripe order. An unusual interplay between spin order and bulk superconductivity was also observed. With increasing pressure the spin order temperature and the size of the ordered moment are not changing significantly. However, application of hydrostatic pressure leads to a remarkable decrease of the magnetic volume fraction $V_{\rm m}(0)$. Simultaneously, an increase of the SC volume fraction $V_{\rm sc}(0)$ occurs. Furthermore, it was found that $V_{\rm m}(0)$ and $V_{\rm sc}(0)$ at all p are linearly correlated: $V_{\rm m}(0) + V_{\rm sc}(0) \simeq 1$. This is an important new result, indicating that the magnetic fraction in the sample is directly converted to the SC fraction with increasing pressure. The mechanism of this transformation, however, is not clear yet and requires further studies. The present results provide evidence for a competition between bulk superconductivity and static magnetic order in the stripe phase of LBCO-1/8, and that static stripe order and bulk superconductivity occur in mutually exclusive spatial regions. Our findings suggest that a pressure of about 5 GPa would be sufficient to completely suppress the static stripe phase and restore bulk superconductivity in LBCO-1/8.

Acknowledgments

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